1 ENVIRONMENTAL RADIATION AND RADIOACTIVITY

1.1 OVERVIEW

Kevin M. Miller

The Environmental Radiation and Radioactivity program area encompasses a variety of projects principally aimed at collecting and interpreting fundamental data on natural and anthropogenic radionuclides in the environment and the related ionizing radiation field produced. In some cases, the projects lead to associated studies of non-nuclear pollutants. The data collected in EML's programs can be used to model environmental pathways and to estimate the dose and the health impact to humans on a global, regional, or local basis. The hallmark of many of these projects over the years has been a rapid response capability to address important environmental issues that arise within the DOE complex, the United States, or around the globe.

A significant component of this program area includes the operation and maintenance of EML's Global Network, a group of over 115 sampling sites dispersed throughout the world for measuring radioactivity in the air and in precipitation. The Network can assess any new introduction of radioactivity into the environment, including episodic releases associated with clandestine or accidental nuclear weapon detonations as well as with accidents associated with nuclear reactors, the transportation sector, and the launch and reentry of space vehicles containing radioactive materials. Included in the Network are Remote Atmospheric Measurements Systems which feature on-site sample analysis and satellite communication for near real-time assessment of events.

Information from the EML Global Network constitutes a significant portion of the world's surface air database. The Network's records of radionuclide concentrations serve as benchmark data with which to test global circulation models developed for climate investigations. Our experience in sampling and analysis, garnered over four decades of Network operations, and the continued commitment to quality assurance has led to EML being designated as a calibration center for the World Meteorological Organization's Global Atmospheric Watch Program.

In addition to projects relating to atmosphere inventories and deposition to land, EML participates in studies to characterize radionuclide distributions in the Arctic Ocean. This work is part of the Office of Naval Research program to determine if radioactive waste management practices of the Former Soviet Union have potentially compromised fisheries or led to radioactivity levels of concern in the Arctic ecosystem.

Special studies are undertaken at facilities within the DOE complex to evaluate the distribution and cycling of radioactive contaminants. Basic information on the dynamics of the systems under study is required to assess the environmental impacts of remediation strategies and their efficacy in environmental restoration programs. In addition, EML's past work in lake sediment coring as part of localized and regional studies involving radionuclides has lead to spin-off studies that focus on toxic and potentially carcinogenic organic compounds. This work is being performed in conjunction with the U.S. Environmental Protection Agency (EPA) as part of their Dioxin Exposure Initiative and Arctic Contaminant Research Program.

The final element of this program area relates to the assessment of environmental radiation fields produced by both natural background and anthropogenic radionuclides. An ongoing study of external radiation exposure from terrestrial and cosmic sources provides fundamental information for assessing the contributions from nuclear technology as well as for setting release criteria for residual radioactivity at sites undergoing environmental restoration. Quality assessment projects that are international in scope are performed in the area of environmental radiation dosimeters, in particular, thermoluminescence dosimeters. Our program also includes standards work for these devices as they represent a principal means for documenting environmental exposures at DOE and other nuclear facilities.

1.2 GLOBAL RADIONUCLIDE DEPOSITION STUDIES

Matthew A. Monetti, Karin M. Decker, Sylvia Hulse, and William Rivera

EML has been investigating the atmospheric deposition of radioactivity for 40 years, and this scientific pursuit continues to provide information addressing issues of concern within the scientific community. In 1958, the laboratory instituted a global network of sampling sites to determine the global transport and fate of radionuclides released as a result of atmospheric testing of nuclear weapons. A single radiologically important fission product, ^{90}Sr, was measured in the samples collected during the first 32 years of the program. The database generated proved valuable for determining factors controlling the global distribution of radioactive fallout, as well as providing source-term information for studies using anthropogenic radionuclides as biogeochemical tracers of various processes. Since the last atmospheric weapons test in 1980, there has been only one reported large-scale release of anthropogenic radioactivity into the atmosphere, namely the Chernobyl Accident. The deposition of ^{90}Sr was insignificant and primarily undetectable from the early 1980s until 1990 except for 1986, the year of the Chernobyl Accident (Monetti and Larsen, 1991; Monetti, in press). As a result of the lack of any definitive atmospheric source of ^{90}Sr and studies conducted during the past two years, the radionuclide analysis of the deposition samples was changed to gamma spectrometry. The radionuclides of particular interest include naturally-produced isotopes, ^{7}Be and ^{210}Pb, and fission products, ^{137}Cs, ^{95}Zr and ^{144}Ce. Gamma spectrometric measurements of fission products enable the program to continue to be used to identify accidental or intentional atmospheric releases of radioactivity. The ^{7}Be and ^{210}Pb data is of special interest to researchers developing global circulation models since it may be used to verify the models. The data will also be available to calculate the source term and inventories of ^{210}Pb and ^{7}Be for colleagues interested in using these radionuclides as biogeochemical tracers. Since the gamma analysis is non-destructive, the samples are archived or used for additional studies. The data generated in these studies can be directly coupled to those found in the Surface Air Sampling Program (SASP) (see Summary No. 1.3). The integration of the results from these two programs leads to the investigation of the removal processes of these radionuclides from the atmosphere.

At the end of 1995, deposition samples were being collected at the 79 locations shown in Figure 1.1. The sampling station in Columbia, Missouri, was removed from the global network as a result of staffing changes at that location. Monthly samples are collected at most of the sites using either an ion-exchange column, a stainless steel pot or a polyethylene bucket. However, samples from

the Australian sites and six sites managed by the United Kingdom's Atomic Energy Authority are collected on a quarterly basis. All of the monthly collections, except for those collected at eight specific locations, are composited into quarterly samples at EML. The samples collected at the other eight sites are analyzed as monthly samples in order to obtain more detailed results of the radionuclide deposition, particularly the short-lived ones. The samples are analyzed by gamma spectrometry and then used for other studies or archived for potential future use. During 1995, these eight sites included Yap Island; Cold Bay, Alaska; Munich, Germany; Columbia, Missouri; Keflavik, Iceland; Bangkok, Thailand; Harare, Zimbabwe; and Guayaquil, Ecuador. These sites were chosen to represent various climatological conditions and a wide global distribution. Special studies of radionuclide deposition were also conducted in 1995 in response to the detonations of underground nuclear weapons by China and France (see Summary No. 1.5).

About 200 samples were processed and analyzed by gamma spectrometry during the past year. Over a quarter of these samples was collected during the Chinese and French testing. These samples were returned to EML by express shipping services to provide us with the opportunity to determine if any short-lived radionuclides were present . The remainder of the analyzed samples was collected as part of the normal studies during 1995. Fission products were below the detection limit in all but four samples. The activity in these four samples was low and temporally and spatially isolated, so they are not likely to represent an unannounced release of anthropogenic radionuclides. The only fission product detected was long-lived ^{137}Cs. Its presence may have been due to resuspension. The natural radionuclides, ^{7}Be and ^{210}Pb, were detected much more frequently, but not in all of the samples. ^{7}Be has a short half-life (53.3 days), so it will only be found in samples analyzed soon after collection. Detectable activities of ^{7}Be were found in 67 samples. Many of the samples analyzed within a few months of collection did not have detectable ^{7}Be activities. ^{210}Pb, however, has a much longer half-life (22.3 years), and this isotope should remain in all the samples that were analyzed. The detector background and matrix blank of ^{210}Pb is a more significant concern for ^{210}Pb measurements. These factors cause the limit of detection for ^{210}Pb and the error associated with ^{210}Pb analysis to be higher. ^{210}Pb was detected in 110 of the samples. Radionuclide deposition based on samples analyzed to date was found to go up to values of 850 and 65 Bq/m^2 per quarter for ^{7}Be and ^{210}Pb, respectively.

The current database of gamma-emitting radionuclide deposition is rather limited, so it is not easily possible to discern any temporal or global trends in the deposition. It is expected that continued studies will reveal these trends. In 1996, attempts will be made to better coordinate these studies with the SASP in order to address some factors involved in the transport and removal of these radionuclides from the atmosphere.

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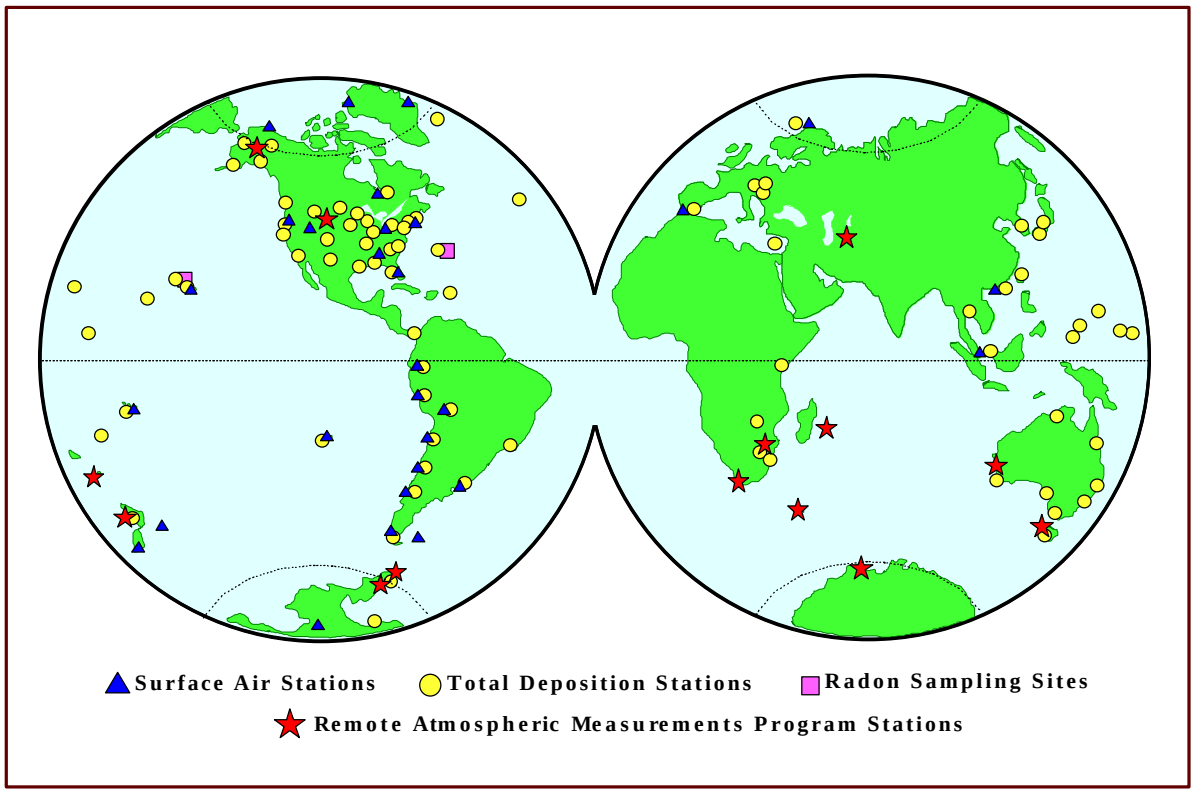

Figure 1.1 EML's Global Sampling Network.

1.3 THE SURFACE AIR SAMPLING PROGRAM (SASP)

John Kada, Colin G. Sanderson, Richard Godwin and William Rivera

Through the Surface Air Sampling Program (SASP), EML operates a network of high volume aerosol samplers at 41 sites around the world. In its early years, the objective of the SASP program was to trace the atmospheric dispersion of radionuclides produced by surface testing of nuclear weapons. SASP also has provided the capability to monitor anthropogenic releases of anthropogenic radionuclides to the atmosphere from accidents, as demonstrated after the 1986 accident at the Chernobyl power reactor and the 1993 accident at the Tomsk-7 facility. In 1995, public concern over a series of subsurface nuclear weapons tests conducted by France at its test sites at Mururoa and Fangataufa Atolls in the Pacific Ocean led to enhanced sampling at several SASP sites in and around the southern Pacific Ocean (see Summary No. 1.5).

The SASP network also provides a platform for the long-term study of the global tropospheric concentration of a number of substances besides artificial radionuclides, and this has become an increasingly important aspect of the SASP program in recent years. SASP regularly measures and reports surface air concentrations of the naturally-produced radionuclides ^{7}Be and ^{210}Pb at all its sites. This data is now widely used in the global climate modeling community to evaluate aerosol transport and scavenging components of global climate models. The recent report of the SASP program (Larsen et al., 1995) adds to this global database of ^{210}Pb and ^{7}Be and includes concentration data for four SASP sites added to the network in 1993: Gibraltar, Singapore, Hong Kong, and Tromso.

The SASP sampling network also serves as a platform for the study of the atmospheric cycles of stable chemical species of interest to scientists involved in climate change and global biogeochemical research. The University of Miami (UOM) analyzes air filter samples from SASP sites in remote regions for a suite of stable chemical species as part of its research on global scale chemistry of the troposphere. Particular emphasis is placed on nitrogen and sulfur species since aspects of the atmospheric chemistry of these species are still not fully understood. Temporal and spatial variations in sulfur and nitrogen concentration in remote areas where anthropogenic influences are minimal provide an opportunity to focus on the natural production, transformation and removal processes affecting these species. ^{210}Pb and ^{7}Be measurements made by SASP at these sites play an important role in the interpretation of the nitrogen and sulfur data by providing clues which help to unravel the effects of transport and aerosol scavenging processes. Because of the important role atmospheric sulfur species are thought to have on the radiative balance of the atmosphere via the “direct” and “indirect” aerosol forcing effects of sulfate aerosol, understanding the atmospheric cycle of these species is also relevant to global climate studies.

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1.4 REMOTE ATMOSPHERIC MEASUREMENTS PROGRAM (RAMP)

Colin G. Sanderson, Steven Minick, Karin M. Decker, Norman Latner, Norman Chiu, Vincent C. Negro, Scott Wurms, Camille Marinetti and John Kada

RAMP is an extension and modification of the Surface Air Sampling Program (SASP) (see Summary No. 1.3) and was initiated in 1987. In addition to having the goals of SASP, RAMP, depending upon the communication system being used, provides real-time and near real-time measurements of gamma-ray emitting radionuclides from remote or weathered-in regions around the world (Sanderson et al., 1994). Thus, radioisotopes having short half-lives, which normally would decay during the period between collection and analysis at EML, are easily measured. In addition, receiving data on a near real-time basis provides EML with a rapid response capability in the event of a nuclear accident.

In a collaborative effort with EML, the University of Miami also analyzes selected RAMP filters for sulfur and nitrogen species which play an important role in aerosol chemistry and may also impact on aerosol-related climate processes. These studies continue to increase the understanding of the atmospheric aerosol which may have a significant impact on the cloud microphysics, precipitation chemistry, and the radiation balance of the entire Antarctic Region and the Southern Oceans.

To accomplish these objectives, RAMP sites (see Figure 1.1) are equipped with remote atmospheric measurements systems (RAMS) which measure the gamma-ray activity in air filter samples on site using either a sodium iodide detector or a mechanically-cooled germanium detector. The resulting spectra are transmitted to the ARGOS communication system flown aboard the National Oceanic and Atmospheric Administration (NOAA) satellites or high through-put geostationary satellites (METEOSAT), transferred to ground stations, and automatically recovered via a telephone link by EML's computer. The spectra are then automatically reconstructed and analyzed at EML. The data are available within 24 hours after field analysis. The analysis system automatically checks data validity and notifies responsible personnel of suspicious analysis results via voice and e-mail, and it maintains a complete current backup set of data and software at all times.

In April 1995, a sodium iodide RAMS was installed near Pinedale, WY. This system, similar to the one installed in Alaska in 1994, utilizes land telephone lines for two-way communications between the remote site and EML. Personnel located at EML in New York can now perform the same analysis functions with the RAMS that site personnel perform via these telephone lines.

A new, completely automatic RAMS (AUTORAMP) has completed its laboratory testing at EML. Unattended operation of EML's RAMS is now possible for one month if daily samples are collected, or up to three months if samples are collected on a three-day cycle. With this new system, the gamma-ray detector is a mechanically-cooled high-purity germanium diode coupled to a portable multichannel analyzer (MCA). A portable computer is used to control the MCA, a "pick & place" robot system, and data flow. Sealed lead acid gel-cell batteries and trickle chargers are used for continuous d.c. current operation. The transmitted spectra and on site peak search analysis are reconstructed and reviewed at EML.

On July 5, 1995, the AUTORAMP (see Figure 1.2) was installed at McClellan AFB, Sacramento, CA, for field testing and evaluation. Although the mechanical functions of the AUTORAMP were operational on July 13, the electrically cooled germanium detector failed. In mid August, another detector was installed and backup sensor switches were added to the system. On August 10, 1995, AUTORAMP became completely operational and began transmitting gamma-ray spectra to headquarters. The AUTORAMP continued to operate flawlessly throughout the test period. Gamma-ray spectra were transmitted daily and analyzed at EML. The field test at McClellan AFB was extended to February 4, 1996.

Future RAMP research will focus on high speed data transmission and analysis. The use of INMARSAT-M satellites or INTERNET e-mail for two -way communications will provide EML with the ability to control or modify from NY all the operations of the RAMS at remote locations and to recover gamma-ray data instantaneously on demand.

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Figure 1.2 Automatic RAMP System.

1.5 MONITORING FOR ATMOSPHERIC RELEASE DURING CHINESE AND FRENCH UNDERGROUND NUCLEAR TESTS

Matthew A. Monetti, John Kada, Richard J. Larsen, Hsi-Na Lee, Colin G. Sanderson, Karin M. Decker, William Rivera, and Kevin J. Clancy

During 1995, both China and France conducted underground nuclear weapons detonations to test recent developments in their nuclear programs, although these tests were formally criticized by other nuclear nations as being unnecessary. The Chinese government conducted a single test on August 17, 1995 at their Lop Nor test site in north-western China. Between September 1995 and February 1996, France performed a series of six underground nuclear tests at its test sites at Mururoa and Fangataufa Atolls in the Pacific Ocean. In spite of reassurances given by the Chinese and French government, the tests evoked widespread expressions of public concern throughout the regions, as well as the world, in part because of fears that anthropogenic radioactivity would be released into the environment.

To help provide objective data relevant to these concerns, EML increased its monitoring of radionuclide activities in surface aerosol and precipitation at sampling sites it maintains under its Global Fallout, SASP, and RAMP programs (see Summary Nos. 1.2, 1.3 and 1.4). The sites utilized to investigate potential releases from the Chinese test were: Bangkok (14° N, 101° E), Chiang-Mai (19° N, 99° E), Taipei (25° N, 122° E), Seoul (37° N, 127° E) and Hong Kong (22° N, 114° E). Those involved in the Pacific region during the French tests were: Norfolk Island (29° S, 168° E), Wellington (41° S, 175° E), Easter Island (27° S, 109° W), American Samoa (14° S, 170° W), Lima, Peru (12° S, 77° W), and Chacaltaya, Bolivia (16° S, 68° W). Our sampling procedures were modified to increase the sampling frequency and to hasten the return and analysis of the samples. Additionally, forward trajectory analyses were performed following each test to predict the transport of radioactivity that may have been released.

Following the Chinese test, weekly deposition samples were collected at Bangkok and Chiang-Mai for four weeks, and at Taipei, Hong Kong and Seoul deposition samples were collected for the months of August and September. All the samples were sent to EML and analyzed as quickly as was possible. In all, 14 deposition samples were analyzed by gamma spectrometry to investigate possible radioactive releases following the Chinese test. During the period of French testing, all three of EML's global network programs were utilized. On-site gamma spectral analysis of weekly air filter samples at the Norfolk and Wellington sites was performed using EML's RAMP system. Air filter samples collected at Easter Island, American Samoa and the South American sites were sent to EML by express shipments and analyzed by gamma spectrometry. Deposition samples were collected at Easter Island, American Samoa and Chacaltaya at weekly intervals instead of the routine monthly sampling protocol. These samples were also quickly delivered to EML for gamma spectrometric analysis. The 164 samples analyzed for the French testing study include 44 RAMP samples, 69 SASP aerosol samples and 51 deposition samples.

Results of the gamma spectrometric analyses of these samples did not indicate that any radioactive material had been released to the atmosphere by the Chinese and French underground tests. If, in the unlikely event there was an atmospheric release of radioactivity, these results would

suggest that it was rather limited in quantity and/or distribution. These special studies have now been discontinued since no further testing is expected, and the sampling sites have resumed the normal operating procedures. The samples collected for these studies are being archived and will be available should any interest in follow-up studies develop.

1.6 THE GLOBAL ATMOSPHERIC WATCH: EML'S ROLE IN QUALITY ASSURANCE

Richard J. Larsen, Philip W. Krey and Merrill Heit

The Environmental Measurements Laboratory has been identified as the Calibration Center for Radioactivity/Quality Assurance Science Activity Center (QA/SAC) for the World Meteorological Organization's (WMO) Global Atmospheric Watch (GAW) Program. In this role, EML will provide direction to GAW in measuring naturally-occurring radionuclides, specifically ^{7}Be, ^{210}Pb, ^{222}Rn and ^{222}Rn progeny. Quality assurance issues related to the sampling and analyses of these isotopes, such as sampler volume determination, filter paper efficiency, and gamma-ray spectrometric analyses, will be identified and guidance will be provided.

Philip W. Krey, Acting Director of EML, is a member of the Principals' Committee that oversees the QA effort for GAW. In October 1995, Mr. Krey gave a poster presentation on "EML's Global Network and Planned Role in GAW" at the First Dual Conference of the WMO/GAW and the International Global Atmospheric Chemistry (IGAC) on the Measurement and Assessment of Atmospheric Composition Change held in Beijing, China. During the conference, Dr. Ernst Brunke from the Atmospheric Trace Gas Research Station, Cape Point, South Africa, and Professor Zhongxiang Hong, Director of the Institute of Atmospheric Physics of the Chinese Academy of Sciences, each requested EML radon analyzers for installation at their measurement stations. Following the conference, Mr. Krey visited the newly established Chinese Global Atmospheric Watch Baseline Observatory (CGAWBO) at Mount Waliguan. During the visit, Dr. Tong Jie, Deputy Director of the Observatory, expressed interest in obtaining a Remote Atmospheric Measurements System (RAMS) from EML (see Summary No. 1.4). EML is currently evaluating the deployment of its radon and RAMS systems.

1.7 RADIONUCLIDES IN THE ARCTIC OCEAN

Thomas M. Beasley

During 1995, samples collected from the Trans-Arctic Ocean Section (TAOS) cruise of 1994 were analyzed for ^{129}I (water) and plutonium isotopes (sediment). In addition, ^{129}I was determined in water samples collected in 1986 from the Beaufort Sea near the coast of Alaska. The purpose of the analyses was to further characterize the distribution of these radionuclides in the Arctic Ocean as part of the Office of Naval Research's Arctic Nuclear Waste Assessment Program (ANWAP).

Iodine-129 in Arctic Ocean Waters. Figure 1.3 shows the results of the ^{129}I analyses from the TAOS collections. At each station, ^{129}I concentrations are maximal at shallow depths ($\leq$ 100 m); at none of the locations do ^{129}I concentrations reach those measured in samples collected from the Barents, Kara, and Laptev Seas during the period 1992-1993. They are more comparable to those measured in the Canadian Basin from collections made in 1993 and 1994 (Beasley, 1995).

In contrast to the TAOS samples, waters collected near the Alaskan Coast in October, 1986 (Beaufort Sea) show: (1) a subsurface maximum in ^{129}I between 300-400 m (Figure 1.4); and (2) elevated ^{129}I at the surface most probably due to the Chernobyl accident in late April of that year. The inventory of ^{129}I from 0-1500 m (3.7×10^{13} atoms m^{-2}) is estimated to be more than an order of magnitude lower than inventories, over comparable depths, in 1993-1994.

If ascribed exclusively to fuel reprocessing releases at Sellafield (U.K.), the 1986 inventory would, at most, double that derived from natural ^{129}I production and above-ground nuclear weapons tests. Because discharges from Sellafield have introduced considerable amounts of ^{137}Cs and other radionuclides to the Arctic Ocean (Cochran et al., 1995; Kershaw and Baxter, 1995 and references therein), ^{137}Cs measurements are in progress to confirm Sellafield as the source of the ^{129}I in these waters.

Plutonium Isotopes in Arctic Basin Sediments. The analysis of surface sediments collected on the TAOS cruise has confirmed the presence of low-ratio Pu throughout the Arctic Ocean basins (Beasley, 1995). Figure 1.5 shows collection sites and the measured $^{240}Pu/^{239}Pu$ sedimentary ratios. Where measurable, $^{241}Pu/^{239}Pu$ in these sediments indicate that in the basin interiors, the "age" of the Pu (i.e., its production age) is near 1957.

The absence of low-ratio Pu in the Greenland ice sheets between 1956 and 1965 (Koide et al., 1985) and the correspondence between the $^{240}Pu/^{239}Pu$ ratios measured there (0.17-0.28) with those measured in soils above 55^0 N in 1970 (0.18-0.19; Krey et al., 1976) argue against weapons-test debris as the source of this Pu. Sellafield discharges in the period 1958-1965 did contain Pu whose $^{240}Pu/^{239}Pu$ ratios ranged between 0.06 and 0.08 (Kershaw et al., 1995), but these ratios increased steadily through 1983 reaching values as high as 0.25. The presence of Sellafield-derived ^{129}I throughout much of the Arctic Ocean Basin would argue against the exclusive input and sedimentation of early Sellafield wastes (low-ratio Pu) to the exclusion of those released during the 1970s and 1980s (high-ratio Pu), particularly when transit times from the Irish Sea to the Barents and Kara Seas can be accomplished within a decade or less (Livingston, 1988; Smith et al., 1990). Thus,

the origin of the low-ratio Pu in sediments as far west as the Canadian Basin suggests a more localized source in the Arctic itself.

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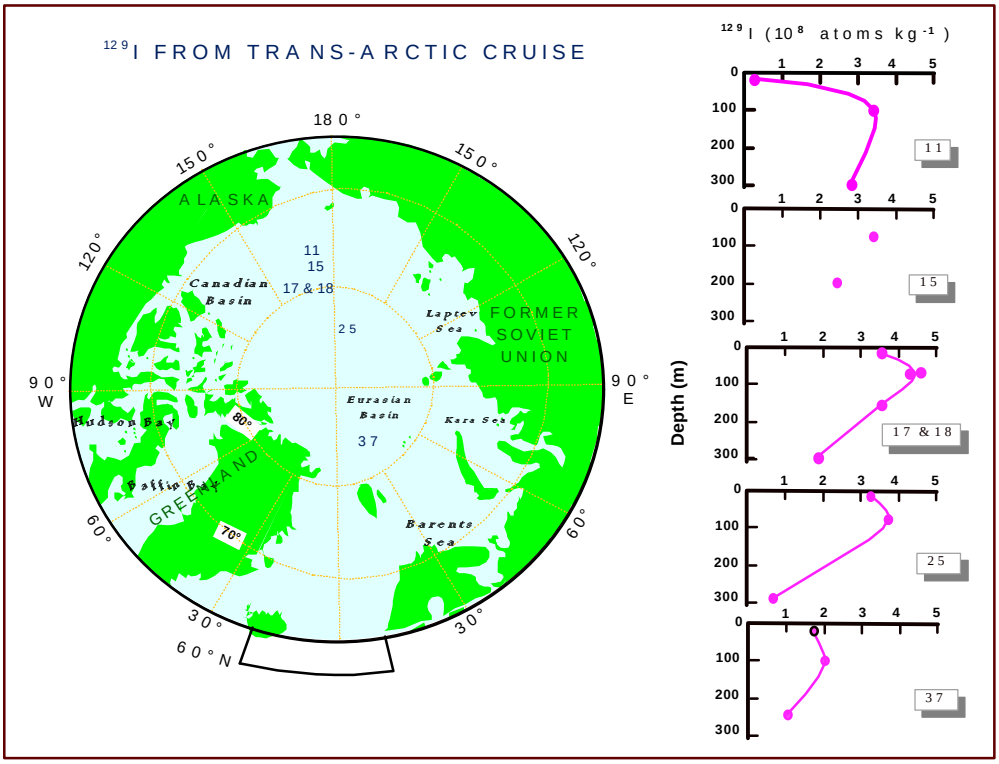


Figure 1.3 Trans-Arctic cruise sampling sites showing depth distributions of ^{129}I.

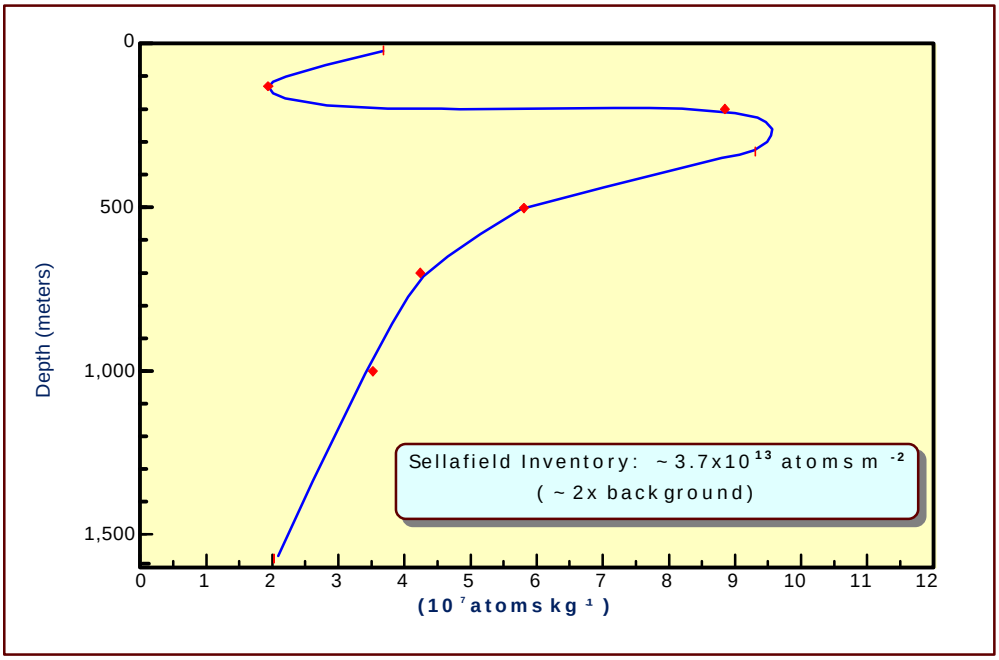
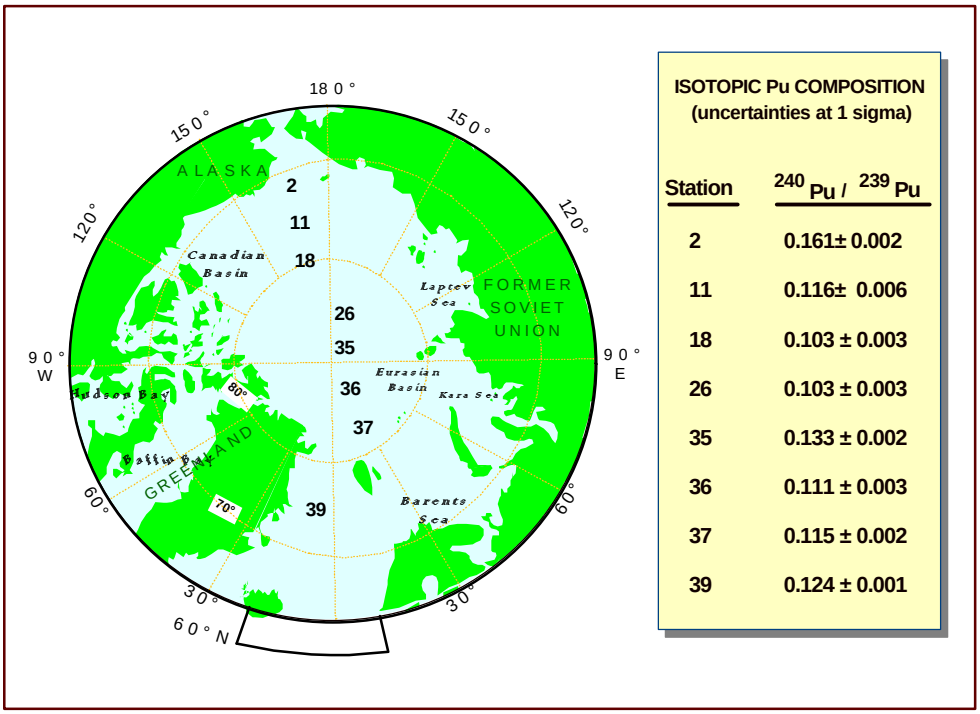


Figure 1.4 Profile of ^{129}I in waters from the Beaufort Sea collected in 1986. Elevated concentrations at the surface indicate input from Chernobyl accident.



ISOTOPIC Pu COMPOSITION
(uncertainties at 1 sigma)

Station	^{240}Pu / ^{239}Pu
2	0.161± 0.002
11	0.116± 0.006
18	0.103 ± 0.003
26	0.103 ± 0.003
35	0.133 ± 0.002
36	0.111 ± 0.003
37	0.115 ± 0.002
39	0.124 ± 0.001

Figure 1.5 Plutonium isotope ratios in samples collected during Trans-Arctic Cruise showing wide spread distribution of low-ratio Pu throughout Arctic Ocean basins.

1.8 PLUTONIUM ISOTOPES IN SOILS AT THE IDAHO NATIONAL ENGINEERING LABORATORY (INEL)

Thomas M. Beasley

During 1995, soil core collections at the Idaho National Engineering Laboratory (INEL) were completed as part of a study to determine the dispersion of Pu away from the Radioactive Waste Management Complex (RWMC). A total of 100 cores (0-10 cm depth) have been blended, tested for homogeneity, and analyzed for gamma-emitting radionuclides. Approximately 30 cores were taken outside of the INEL site boundaries; the remaining cores were collected at various distances from the RWMC (Beasley, 1995). Subsamples of all cores have been processed for mass spectrometry measurements of Pu isotopes and ^{237}Np; approximately 50% of the analyses has now been completed.

Figure 1.6 shows representative $^{240}Pu/^{239}Pu$ ratios for cores collected in the direction of the principal winds at the site. Global fallout values for the $^{240}Pu/^{239}Pu$ ratio at the INEL are expected to lie within the range determined by Krey et al. (1976), namely 0.176 ± 0.014. From the data shown in Figure 1.6, it is clear that substantially lower ratios are observed in several of the cores collected southwest of the RWMC. For example, assigning a $^{240}Pu/^{239}Pu$ ratio of 0.05 to RWMC Pu (a value typical of Pu processed at Rocky Flats, CO) the amount of RWMC-derived Pu at coring site 56 is

estimated to be ~ 40 percent of the measured 70 ± 1 Bq m^{-2} of total ^{239}Pu + ^{240}Pu at that location.

Because winds can reach velocities > 100 km hr^{-1} at the INEL, redistribution of surface soils is constantly taking place. Consequently, Pu released from the RWMC in the late 1960s and early 1970s can be transported to great distances. Equally probable, however, is that soils once contaminated with RWMC-derived Pu can be winnowed over time and replaced or mixed with soil carrying only a fallout Pu signature. It is not surprising, therefore, that in the intervening three decades between Pu release at the RWMC and our soil sampling, redistribution processes have operated to blur what may have initially been a more easily identifiable "footprint" of the Pu released at that facility.

The Idaho Chemical Processing Plant (ICPP) has processed highly-enriched uranium (HEU) over its operating history (maximum percent ^{235}U = 100; Benedict et al., 1981). Thermal neutron irradiation of ^{235}U produces ^{236}U and ^{237}Np. We analyzed core 54, collected near the ICPP, for ^{236}U and find atom concentrations of (7.8 ± 1.1) x 10^{8} atoms g^{-1} soil and a $^{235}U/^{238}$ atom ratio of 0.007296 ± 0.000011, a value that exceeds that of natural uranium, i.e., 0.00725. Because ^{236}U has not been reported as measurable in global fallout, its presence at site 54 must result from stack emissions at the ICPP. Moreover, the concentration of ^{237}Np at site 54 (~ 2 x 10^{8} atoms g^{-1} soil) is elevated (30-40%) with respect to those we have measured in other cores to date.

We have confirmed the presence of both ^{236}U and ^{237}Np in the ICPP waste stream by analyzing water samples from monitoring wells in the Snake River Plain aquifer downgradient from this facility. Near the ICPP, ^{236}U and ^{237}Np concentrations were 3 x 10^{11} and 1 x 10^{9} atoms 1^{-1}, respectively. Uranium-236 could be traced to distances of ~10 km while ^{237}Np was measurable only within 2 km of the plant. We plan further core analyses for ^{236}U to determine the extent to which it can be detected across the INEL site.

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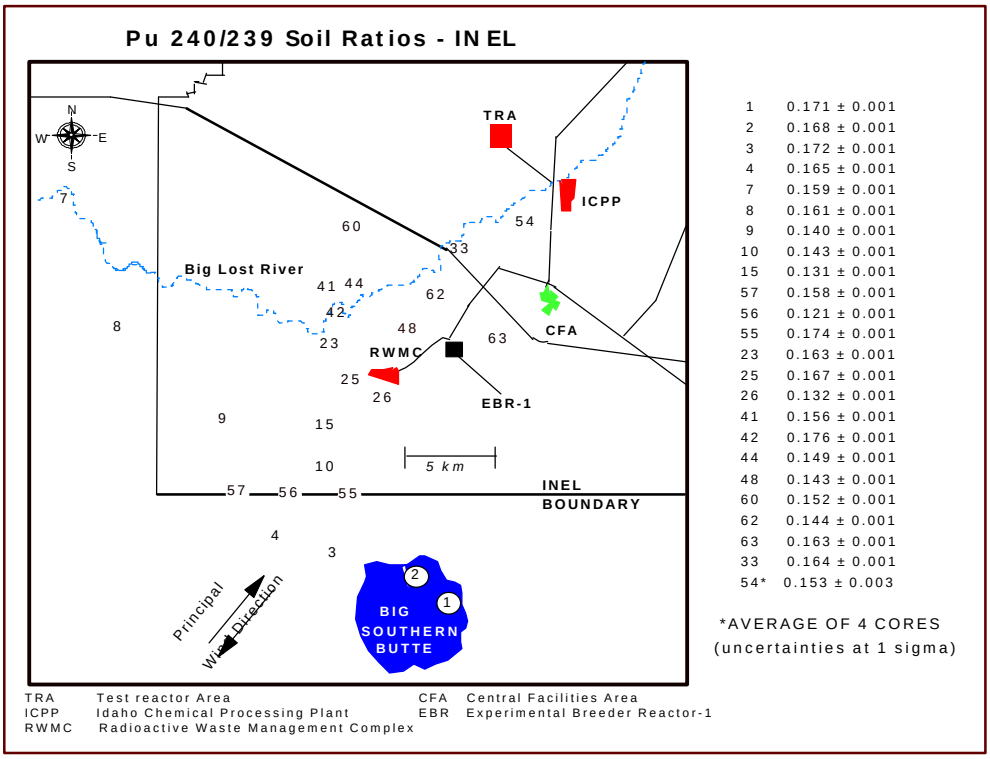

Figure 1.6 Typical $^{240}Pu/^{239}Pu$ ratios in soils at the Idaho National Engineering Laboratory.

1.9 HISTORY OF DIOXIN-LIKE COMPOUNDS IN LAKE SEDIMENT CORES

Matthew A. Monetti, John Kada and Merrill Heit

For this study, EML has collaborated with the EPA's National Center for Environmental Assessment in Washington, D.C., to examine the chronological record of deposition of dioxin-like compounds, i.e., dioxins, furans and coplanar polychlorinated biphenyls (PCBs), revealed in sediments from lakes located in the contiguous United States and Alaska. Since these compounds are highly toxic and are likely to be human carcinogens, the EPA, as part of their Dioxin Exposure Initiative, has an interest in determining the temporal trends in the levels, patterns and profiles of dioxins, furans and PCBs. EML has been involved with these types of investigations since the 1970s. Over the past twenty years, EML has collected sediment cores from 64 lakes or reservoirs in the U.S. and an archive of these samples has been maintained at the Laboratory.

Sediment cores from 11 lakes (Figure 1.7) were chosen for investigation in this study based on certain criteria, including distance from known sources of dioxin-like compounds and lack of point-sources of dioxin-like compounds in the watershed and regional representative lakes having good sediment-dating resolution in the time-period of interest (back to 1930 or further). As a result of previous studies, data was previously available for ^{137}Cs and, in some instances, excess ^{210}Pb, which was used to establish the sedimentation rates and chronology for the cores. Samples representing ten

year intervals were prepared from the cores. Some samples, particularly those from pre-industrial sections of the core, represented larger time periods. Eighty samples (including three blind replicates) were analyzed for dioxins, furans and PCBs by a contractor laboratory selected by the EPA. The contractor followed EPA Method 1613 which uses high resolution gas chromatography / high resolution mass spectrometry techniques. Results included concentrations of total congener group for tetra through octa-chlorinated dioxins and furans, in addition to several 2,3,7,8 -substituted isomers and several PCBs (77, 118, 105, 126, 156 and 169). The database is likely to be the largest of its kind, containing 2,464 values for dioxin-like compounds.

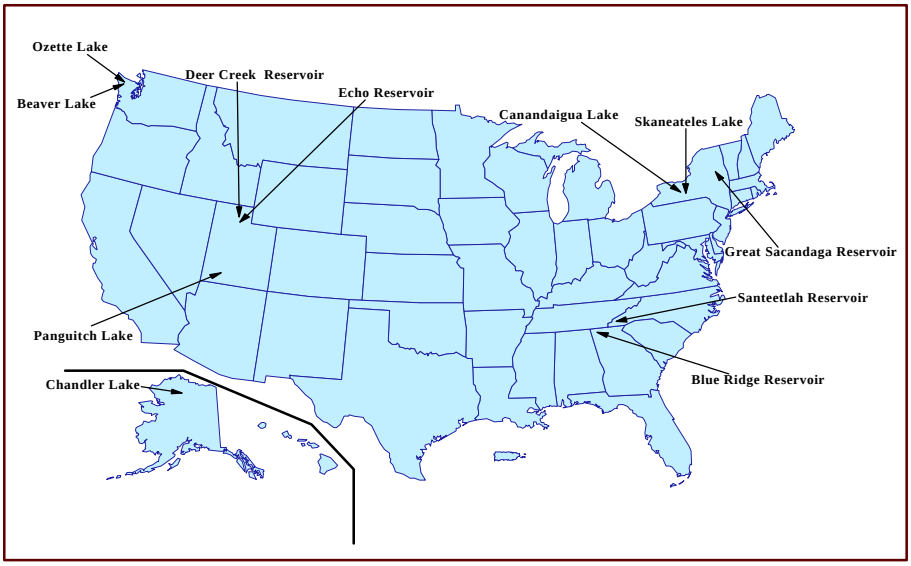

Figure 1.7 Sediment core sample locations.

1.10 ENVIRONMENTAL RADIATION MEASUREMENTS USING PRESSURIZED IONIZATION CHAMBERS (PICs)

Peter Shebell, Kevin M. Miller, William Van Steveninck,
Vincent C. Negro, and Gladys A. Klemic

Routine monitoring of the penetrating component of the environmental radiation field (gamma plus cosmic) using pressurized ionization chambers continued at our regional baseline station in Chester, NJ. Additional monitoring is done at Princeton, NJ. This study has been part of a long-term program whose goal is to document the natural variations as well as any anthropogenic perturbations in the natural background.

A comprehensive analysis of the eighteen year record of daily average dose rates at our Chester station was completed this year. The focus of the study was a quantitative description of the temporal variations that result primarily from snow cover, soil moisture, and atmospheric scavenging of radon progeny. Figure 1.8 is a plot of the eighteen year record illustrating the overall magnitude and daily variations in the dose rate. These results were summarized in a paper presented at the Natural Radiation Environment VI Symposium, Montreal, Canada (Shebell and Miller, in press). The paper also provided means and extremes of the entire record. An analysis of the data by season indicated that the winter months exhibited the largest variation and lowest dose rate. Frequency histograms were generated to show the variations in the daily average dose rate.

The analysis and summary of the eighteen year database provided a natural conclusion to routine monitoring at Chester. A decline in, and a reallocation of resources, led to the decision to terminate the program at Princeton as well. Regional environmental monitoring with PICs will probably continue in a limited capacity.

Monitoring began at Brookhaven National Laboratory (BNL) as part of the 11th International Intercomparison of Environmental Dosimeters. The initial 42-day record of one-minute exposure rates showed a brief but dramatic increase in the exposure rate at noon on the 10th of December. The elevation lasted for only two consecutive one minute measurements and was about six times the 42-day average. It has not been linked to any activities or known releases at BNL. The possibility of equipment malfunction has not been eliminated. An additional system has been deployed to further investigate the situation, as well as to provide a backup unit.

Reference

Shebell, P. and K. M. Miller
"Analysis of Eighteen Years of Environmental Radiation Monitoring Data"
Environment International, in press

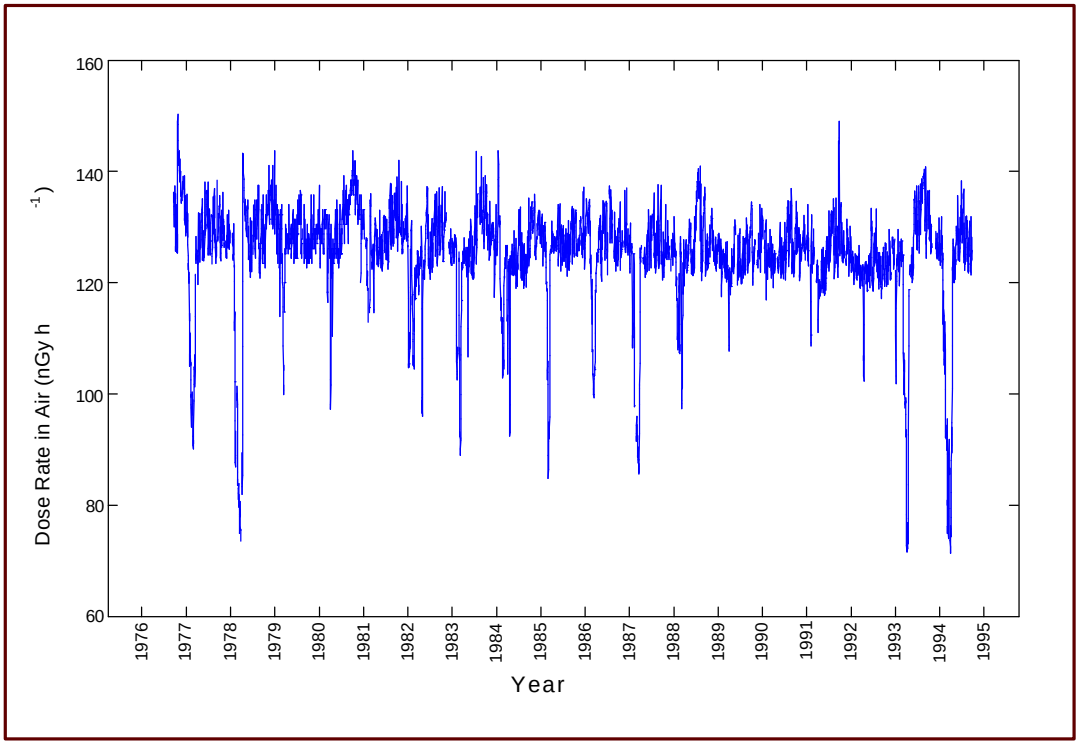

Figure 1.8 Average daily dose rate in air at EML's regional baseline station in Chester, NJ, from 9/1/76 - 9/1/94.

1.11 ENVIRONMENTAL THERMOLUMINESCENCE DOSIMETRY

Gladys A. Klemic and Nestor Azziz

EML's thermoluminescence dosimetry (TLD) program includes maintaining state-of-the-art instrumentation for routine environmental radiation monitoring, the planning and implementation of international intercomparisons, contributions to the development of national standards, special field applications (see Summary Nos., 2.6, 2.7 and 6.18) and investigations of the use of TLDs in mixed gamma-neutron radiation fields (see Summary No. 2.8).

EML maintains two TLD readers in addition to the EML-designed reader that has been in use since 1974. An automated hot gas reader is used for routine measurements and large scale applications, including quality control for the international intercomparisons and batch testing of chips. A commercially available manually operated, linearly heated planchet reader is being used for research applications, including glow curve analysis. In 1995, EML's ^{137}Cs calibration facility was upgraded to include improved sample positioning and the use of recalibrated Shonka-Wyckoff chambers to provide traceability to the National Institute of Standards and Technology (NIST).

International Intercomparisons

The International Intercomparisons of Environmental Dosimeters were initiated in 1974 to assess the performance of passive, integrating detectors in the measurement of environmental radiation and to identify and investigate special problems associated with such measurements. These intercomparisons are presently the only available large scale and universally recognized quality assurance program for passive environmental dosimetry. Participation in the program is voluntary and results are reported without identifying individual participants.

Invitations for the 11th Intercomparison were mailed in late 1995, and 125 participants from 36 countries have registered to participate. The intercomparison will be held at BNL in collaboration with NIST and Idaho State University. An undisturbed field at BNL was characterized by *in situ* gamma spectrometry and ionization chamber measurements and was selected for the site of field deployment. Routine monitoring with EML's pressurized ionization chamber (see Summary No. 1.10) and TLDs began in December 1995.

National Standards Work

In April 1995, EML hosted a meeting of the American National Standards Institute (ANSI) Working Groups N13.29 and N13.37. ANSI standard N13.37 is expected to replace an earlier document (N545) as the type-testing standard for environmental TLD systems. ANSI standard 13.29 will establish performance criteria for environmental dosimetry processors and will be analogous to a similar standard for personnel dosimetry (N13.11) which is currently the basis for the National Voluntary Laboratory Accreditation Program (NVLAP). The draft of ANSI N13.29 was submitted to the Health Physics Society in late 1995 and is presently under review. Results from the

International Intercomparisons of Environmental Dosimeters have proved useful in helping to set procedures and tolerance levels for these standards.